

(equivalent to the substrate of the claimed invention) and a hard coating applied thereon. The Examiner also states that the hard coating of Kayanoki (equivalent to the coating liquid and the hard coat film of the claimed invention) comprises fine particles of a composite oxide (equivalent to the composite metal oxide particles of the claimed invention) having an average particle diameter in the range of from 1 to 100 nm (thus meeting the particle size limitations) and an epoxy group containing silicon compound (equivalent to the matrix-forming component of the claimed invention). In addition, the Examiner indicates that the composite oxide of Kayanoki is preferably composed of iron oxide, titanium oxide and a silica component, and, further, that the weight ratio of the iron oxide to the titanium oxide is in the range of 0.005 to 1.0, and the weight ratio of silica to the sum of the iron oxide and titanium oxide is in the range of 0.001 to 1.0. The Examiner goes on to state that the fine particles of Kayanoki are surface treated with an organosilicon compound, thus meeting the limitations of claims 3 and 6 of the present invention, and that the hard coating of Kayanoki may be provided with a mono-layered or multi-layered anti-reflection film, thus meeting the limitations of claims 5 and 10-12 of the present invention.

The Examiner concludes that Kayanoki substantially discloses the claimed invention but does not specifically state that the weight ratio of the iron oxide to the titanium oxide may be 0.0005 to less than 0.005. However, the Examiner takes the position that it would have been obvious to one of ordinary skill in the art to have determined the optimum weight ratio of the iron oxide to the titanium oxide through routine experimentation, given that the blend ratio of the iron oxide and the titanium oxide may be varied to obtain shielding within a specific wavelength region, i.e., UV region vs. visible light region, as evidenced by U.S. Patent No. 6,077,341 to Terasse et al.

The Examiner notes that Terasse et al. disclose silica-metal oxide particulate composites wherein the metal oxide particulates are titanium oxide and ferrous or ferric oxide and may be used in combination. Further, the Examiner points out that Terasse et al. teach that the specific metal oxide and the amount of the specific metal oxides are selected depending on the particular purpose of the composite material; for example, UV shielding function can be imparted to the composite material by varying the amount of titanium oxide, zinc oxide and iron oxide. The Examiner concludes that the present application does not have criticality in the claimed ratio.

Applicants argue that the present invention, as recited in the claims, provides a coating liquid for forming a hard coat film, comprising a matrix-forming component and particles of a composite metal oxide, wherein the composite metal oxide particles are composed of an iron oxide component and a titanium oxide component, the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  being in the range of 0.0005 to less than 0.005, provided that  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$  represent the weight in terms of  $\text{Fe}_2\text{O}_3$  of the iron oxide component and the weight in terms of  $\text{TiO}_2$  of the titanium oxide component, respectively, and wherein the composite metal oxide particles have an average particle size ranging from 1 to 100 nm.

The present invention, as recited in the claims, also provides for a coating liquid for forming a hard coat film, comprising a matrix-forming component and particles of a composite metal oxide, wherein the composite metal oxide particles are composed of iron oxide, titanium oxide and silica, the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  being in the range of 0.0005 to less than 0.005, and the weight ratio  $\text{SiO}_2/(\text{Fe}_2\text{O}_3 + \text{TiO}_2)$  being in the range of 0.001 to 1.0, provided that  $\text{Fe}_2\text{O}_3$ ,  $\text{TiO}_2$  and  $\text{SiO}_2$  represent the weight in terms of  $\text{Fe}_2\text{O}_3$  of iron oxide, the weight in terms of  $\text{TiO}_2$  of titanium oxide and the weight in terms of  $\text{SiO}_2$  of silica, respectively, and wherein the composite metal oxide particles have an average particle size ranging from 1 to 100 nm.

Hence, the characteristic feature of the coating liquid for forming a hard coat film according to the present invention resides in that the composite metal oxide particles contained as a coating film component are composed of:

(i) an iron oxide component and a titanium oxide component, the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  being in the range of 0.0005 to less than 0.005, provided that  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$  represent the weight in terms of  $\text{Fe}_2\text{O}_3$  of the iron oxide component and the weight in terms of  $\text{TiO}_2$  of the titanium oxide component, respectively, the composite metal oxide particles having an average particle size ranging from 1 to 100 nm, or

(ii) iron oxide, titanium oxide and silica, the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  being in the range of 0.0005 to less than 0.005, and the weight ratio  $\text{SiO}_2/(\text{Fe}_2\text{O}_3 + \text{TiO}_2)$  being in the range of 0.001 to 1.0, provided that  $\text{Fe}_2\text{O}_3$ ,  $\text{TiO}_2$  and  $\text{SiO}_2$  represent the weight in terms of  $\text{Fe}_2\text{O}_3$  of iron oxide, the weight in terms of  $\text{TiO}_2$  of titanium oxide and the weight in terms of  $\text{SiO}_2$  of silica, respectively, the composite metal oxide particles having an average particle size ranging from 1 to 100 nm.

A coating film component containing the above composite metal oxide particles enables the formation of a hard coat film which is free from being tinted with yellowish hue and also free from exhibiting photochromism and which has satisfactory weather resistance.

As a result, the thus-formed hard coat film has advantageous properties, i.e., being excellent in weather resistance, having a high surface hardness to thereby ensure excellent scuffing resistance and attrition resistance, not exhibiting photochromism so that it is free from discoloration by light irradiation, having high dye affinity and high transparency, having high hardness, and exhibiting not only excellent scuffing resistance but also high refractive index.

Kayanoki discloses a hard coating having a high refractive index which is preferably formed of the following three components: A, B and C.

Component A is sol obtained by dispersing fine particles of at least one oxide selected from iron oxide, titanium oxide, cerium oxide, zirconium oxide, antimony oxide, zinc oxide and tin oxide, a mixture of these, or a composite oxide of these, and which have an average particle diameter in the range of from 1 to 100 nm, in water or other solvent. As the above fine particles, for example, composite oxide particles formed of an iron oxide component, a titanium oxide component and a silica component, or composite oxide particles formed of a cerium oxide (or ceria) component, a titanium oxide component and a silica component are preferred. When the composite oxide particles are formed of an iron oxide component, a titanium oxide component and a silica component, it is preferable that the weight ratio of  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  is in the range of 0.005 to 1.0, and the weight ratio of  $\text{SiO}_2/(\text{Fe}_2\text{O}_3 + \text{TiO}_2)$  is in the range of 0.001 to 1.0, in which the amount of the iron oxide component is calculated as  $\text{Fe}_2\text{O}_3$ , and the amount of the titanium oxide component is calculated as  $\text{TiO}_2$ , and the amount of the silica component is calculated as  $\text{SiO}_2$ . When the composite oxide particles are formed of a cerium oxide component, a titanium oxide component and a silica component, it is preferable that the weight ratio of  $\text{Ce}_2\text{O}_3/\text{TiO}_2$  is in the range of 0.1 to 1.0, and the weight ratio of  $\text{SiO}_2/(\text{Ce}_2\text{O}_3 + \text{TiO}_2)$  is in the range of 0.05 to 0.5, in which the amount of the cerium oxide component is calculated as  $\text{Ce}_2\text{O}_3$ , the amount of the titanium oxide component is calculated as  $\text{TiO}_2$ , and the amount of the silica component is calculated as  $\text{SiO}_2$ .

Component B is an epoxy-group-containing silicon compound and Component C includes 4-hydroxybutyl acrylate and 4-hydroxybutyl methacrylate. (See Kayanoki, col. 7, lines 31-63).

Upon comparison, the present invention is clearly different from the Kayanoki reference in that, if the weights of the iron oxide components and the titanium oxide components are expressed in terms of  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$ , respectively, the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  is in the

range of 0.0005 to less than 0.005 in the present invention, while the weight ratio of Kayanoki is in the range of 0.005 to 0.15. Please note that in the present invention the weight ratio is up to less than 0.005, thus not containing 0.005.

When the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  of the fine compound oxide particles is 0.005 or greater, as in Kayanoki, the formed hard coat film is photochromic, thereby suffering from tone change by ultraviolet irradiation, so that a hard coat film which is excellent in weather resistance, particularly from the viewpoint of not coloring in appearance, cannot be formed. In this regard, reference is made to page 3, line 18 through page 4, line 11 of the present specification:

The hard coat film described in the Japanese Patent Laid-open Publication No. 5(1993)-2102, although improved in weather resistance, may be slightly tinted with yellowish hue because the iron oxide *per se* is yellowish. Further, there is the problem that the above composite metal oxide containing iron oxide is photochromic, so that when the hard coat film containing the composite metal oxide is irradiated with, for example, ultraviolet rays, the tone of the hard coat film *per se* is changed. These coloring and tone changes are generally canceled when the irradiation is terminated, but there are occasions in which the restoration to the original state is difficult. Still further, when an anti-reflective multicoat layer is formed on the above hard coat film, the restoration of the discolored hard coat film to the original tone is inhibited.

By contrast, in the present invention, when the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  of the fine compound oxide particles contained as a coating film component falls within the extremely limited range of 0.0005 to less than 0.005, the hard coat film is not photochromic and is free from turning yellow. Moreover, as long as the weight ratio is as mentioned above, a hard coat film which has not only excellent weather resistance but also high refractive index and high transparency and which is excellent in resistance to hot water, weatherability, scuffing resistance,

attrition resistance and dye affinity and, further, adherence to substrates can be formed. Obtaining the advantageous properties of the present invention simultaneously is a result of the use of a weight ratio in this limited range, and cannot be derived solely from the UV shielding abilities of the components.

U.S. Patent No. 6,077,341 to Terasse et al. is concerned with a composite comprising metal oxide particulates and silica agglomerates having voids formed by random stacking of scaly silica primary particles, and metal oxide particulates supported on the surfaces and the inner surfaces in the voids of the silica agglomerates.

The composite of Terasse et al. is a mixture of metal oxide particulates and silica agglomerates and is not the composite oxide of the present invention.

The Examiner makes reference to the use of mixtures of two metal oxides by Terasse et al. This is not the teaching of the present invention, where a composite oxide is used.

Furthermore, Terasse et al. contains no teaching concerning the range of the ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  or  $\text{SiO}_2/(\text{Fe}_2\text{O}_3 + \text{TiO}_2)$  in composite metal oxide particles. Therefore, it is apparent that Terasse et al. does not teach or suggest a coating liquid for forming a hard coat film which is free from photochromism and has not only excellent weather resistance but also high refractive index and high transparency which is excellent in resistance to hot water, weatherability, scuffing resistance, attrition resistance and dye affinity and, further, adherence to substrates can be exerted by regulating the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  of the fine composite metal oxide particles contained as a coating film component so as to fall within the extremely limited range of 0.0005 to less than 0.005.

Comparison of the results of Example 1 and Comparative Example 1 of the present application shows the effect which is attained by regulating the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$ , wherein  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$  represent the weight in terms of  $\text{Fe}_2\text{O}_3$  of the iron oxide component and

the weight in terms of  $\text{TiO}_2$  of the titanium oxide component of the composite metal oxide particles contained as a coating film component, respectively, so as to fall within the extremely limited range of 0.005 to less than 0.005 according to the present invention.

In Example 1, a hard coat film was produced with the use of a composite metal oxide whose weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  was 0.002 and the properties of the hard coat film were evaluated. In Comparative Example 1, a hard coat film was produced with the use of a composite metal oxide whose weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  was 0.02 and the properties of the hard coat film were evaluated.

TABLE 1

	Sol wt. ratio	Photo chromism	High refractivity	Scuffing Resistance	Appearance	Dye Affinity	Weather resistance		Cloudiness	Stability	
	$\text{Fe}_2\text{O}_3/\text{TiO}_2$						coloring	adherence		25 days	45 days
Example 1	1/499	none	0	A	0	0	none	0	0	0	x
Comp. Ex. 1	1/49	occurred	0	A	0	0	occurred	0	0	0	x

As is apparent from a comparison of the results of Table 1, when the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$ , wherein  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$  represent the weight in terms of  $\text{Fe}_2\text{O}_3$  of the iron oxide component and the weight in terms of  $\text{TiO}_2$  of the titanium oxide component of the composite metal oxide particles contained as a coating film component, respectively, is in the range of 0.0005 to less than 0.005 (see Example 1), a hard coat film which is free from photochromism and excellent in weather resistance can be formed.

By contrast, when the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$  is 0.005 or greater, as in Kayanoki and Terasse et al., the hard coat film has the photochromism and a hard coat film having excellent weather resistance cannot be formed (see Comparative Example 1). The simultaneous optimization of these properties is not taught by Terasse et al., and cannot be predicted on the basis of properties taught by Terasse et al.

In conclusion, it is apparent from the foregoing that none of the cited references discloses regulating the weight ratio  $\text{Fe}_2\text{O}_3/\text{TiO}_2$ , wherein  $\text{Fe}_2\text{O}_3$  and  $\text{TiO}_2$  represent the weight in terms of  $\text{Fe}_2\text{O}_3$  of the iron oxide component and the weight in terms of  $\text{TiO}_2$  of the titanium oxide component of the compound oxide particles contained as a coating film component, respectively, so as to fall within the range of 0.0005 to less than 0.005. In addition, none of the cited references teaches the effect attained by this limited range.

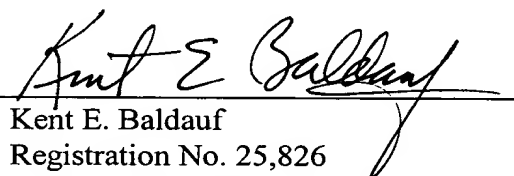
Hence, the present invention is neither taught nor suggested by the cited references.

In light of the above remarks, reconsideration and allowance of claims 1-12 are respectfully requested.

Respectfully submitted,

WEBB ZIESENHEIM LOGSDON  
ORKIN & HANSON, P.C.

By

  
Kent E. Baldauf  
Registration No. 25,826  
Attorney for Applicants  
700 Koppers Building  
436 Seventh Avenue  
Pittsburgh, Pennsylvania 15219-1818  
Telephone: 412-471-8815  
Facsimile: 412-471-4094





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MARKED-UP VERSION OF CHANGES MADE

IN THE SPECIFICATION:

The paragraph beginning on page 1, line 17 has been replaced with the following rewritten paragraph:

Furthermore, the present invention relates to a substrate coated with a hard coat film which is formed from the above coating liquid.

The paragraph beginning on page 3, line 18 has been replaced with the following rewritten paragraph:

However, the hard coat film described in the Japanese Patent Laid-open Publication No. 5(1993)-2102, although improved in weather resistance, may be slightly tinted with a yellowish hue because the iron oxide *per se* is yellowish. Further, there is the problem that the above composite metal oxide containing iron oxide is photochromic, so that[,] when the hard coat film containing the composite metal oxide is irradiated with, for example, ultraviolet rays, the tone of the hard coat film *per se* is changed. These coloring and tone [change] changes are generally canceled when the irradiation is terminated. However, there are occasions in which the restoration to the original state is difficult in films containing photochromic metal oxide [having photochromic].

The paragraph beginning at page 9, line 17 has been replaced with the following rewritten paragraph:

Organosilicon compounds generally known as silane coupling agents are used as the above organosilicon compound, and the appropriate one is selected from among those in conformity with, for example, the types of matrix-forming component and solvent. Examples of the organosilicon compounds include tetraethoxysilane, methyltrimethoxysilane, trimethylchlorosilane, vinyltriethoxysilane,  $\gamma$ -glycidoxypropyltriethoxysilane or  $\gamma$ -glycidoxypropylmethyldiethoxysilane.

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